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Optical transmission enhancement of Fluorine doped Tin Oxide (FTO) on glass for thin film photovoltaic applications

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Abstract

Transparent conductive oxide layers (TCO) are used as electrodes for thin film applications. The optical performance of TCOs can be improved by implementing textured surfaces that will increase the overall transmission of light through the TCO layer. In this paper, a model is developed using Synopsys TCAD to accurately predict the behavior of a thin film of fluorine doped tin oxide (FTO) on a glass substrate. Its accuracy is assessed using experimental data and was further validated by using the simulated transmission curve to estimate the thickness. It is then used to assess the effect of two types of texturization patterns: rectangular and pyramidal. Inductively coupled plasma- reactive ion etching (ICP-RIE) was used to implement rectangular type patterns on FTO samples. The results show that the rectangular patterns result in minor total transmission gains of 1 – 2 percentage points. This is due to a reduced absorption caused by the removal of material. The best pyramidal patterns increased total transmission by over 5 percentage points. These increases are attributed to reduced reflection.

Keywords: TCO; FTO; TCAD; etching; texturing; average transmission.

1. Introduction

In a thin film solar cell, the front electrode is often made up of a transparent conductive oxide layer (TCO). Currently, indium tin oxide (ITO) is the most commonly used TCO; however, long-term use of ITO is in jeopardy due to the scarcity of indium. This scarcity, combined with an ever growing demand for TCOs, has caused the price of ITO to increase rapidly [1]. As a result, many groups are working to find a material which could be a suitable replacement for ITO. Fluorine doped tin oxide (FTO) is a low cost alternative which has the added benefits of being stable at high temperatures and in acidic and hydrogen rich environments. Consequently, FTO is growing in popularity for devices that require high

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temperature or hydrogen rich fabrication environments such as organic light emitting diodes, organic solar cells, inorganics thin film solar cells and dye-sensitized solar cells.

As the TCO acts as an electrode, its electrical properties are critical to its effectiveness; however, because it is the front layer of the cell, the TCO must be highly transparent as well in order to maximize the amount of light reaching the active layers. One of the parameters that affect both the optical and electrical properties of a TCO is its surface roughness. Smooth surfaces lower the contact resistance and also minimize localized field effects which can be important for devices such as organic LEDs [2-4]. Well-designed rough or patterned surfaces can be used to trap incident light, increasing the amount of light which is absorbed by the active layers of a solar cell [5, 6]. The patterned surface can do this by scattering the incoming light, increasing the optical path length of the light within the solar cells. Optimizing these patterns is the subject of this paper.

The purpose of this study was to develop a model which would be able to accurately predict the optical characteristics of a thin FTO layer on glass. This was done with Synopsys TCAD. Using this model, the effect of various etching patterns on the total transmission of light through the sample is then considered.

2. Simulation Structure

Figure 1 represents the structure that was simulated using the Synopsys TCAD tools [7]. The structure consisted of a glass layer on top of an FTO layer. On the left and right of the structure are side contacts which reflected all incident light back into the structure. The contacts above and below the structure are fully transparent allowing all the light to pass through. The perfectly matched layers (PML) at the top and bottom of the structure are composed of silicon with a high extinction coefficient meaning that all light incident on the PML layers is absorbed.

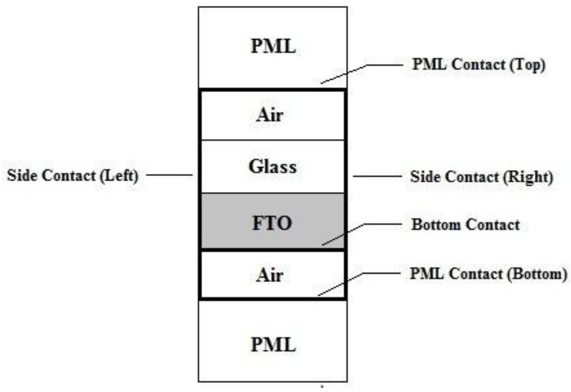


Fig. 1. Cross section of the simulated glass and FTO structure (Figure not to scale).

Figure 2 shows the two types of patterns modeled: one using rectangular features (Fig. 2a) and one using triangular features (Fig. 2b). The width, pitch and etch depth were varied for the rectangular patterns; the opening angle was varied for the pyramidal patterns. In all cases the maximum FTO thickness was held constant at 500 nm. The patterns were implemented at FTO/Air interface. The light source was located in the air layer above the glass layer. The light was uniformly spread over the sample and was perpendicularly incident on the surface.

3. Physics based TCAD model

Synopsys TCAD is a multipurpose software package that contains several different modeling components. In this study, the ray tracer function was used to simulate the optical characteristics of the structures. The model calculates the optical carrier generation rate in each of the PMLs for each wavelength. To do this, it relies on refractive index and extinction coefficient of the materials involved. The FTO data was provided by the supplier while generic glass data was used for the glass layer. The model uses this n and k data to calculate the complex refractive index. The optical carrier generation rate in the top PML layer is used to calculate reflection while that in the lower PML is used to calculate transmission. Finally, the absorption of the structure was calculated by using equation 1,

$$T + R + A = 1 \tag{1}$$

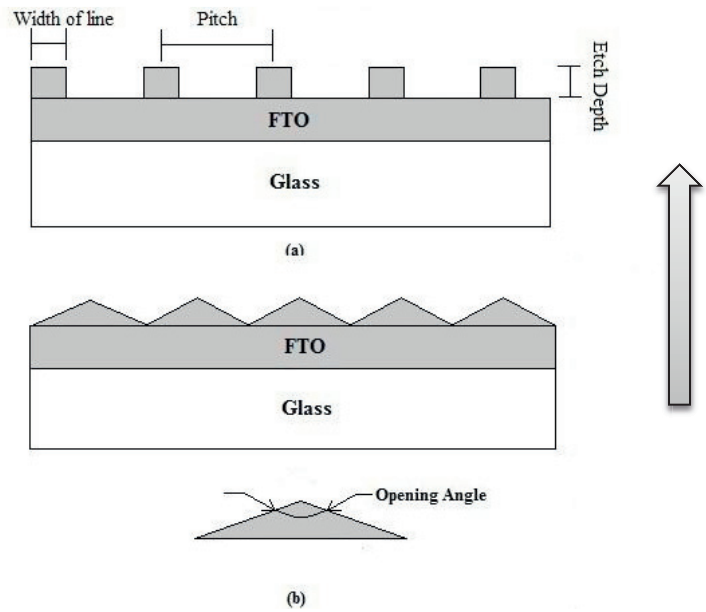


Fig. 2. Cross section of the simulated glass and patterned FTO structure a) Rectangular patterned FTO b) Pyramidal patterned FTO (Figure not to scale) (Arrow indicates the direction of incoming light).

4. Simulation Validation

In order to assess the accuracy of the simulation, the output was compared to experimental results. Figure 3 shows the total transmission of an un-etched FTO sample as simulated with Synopsys TCAD and measured experimentally respectively. The result shows that the simulation follows the same trend as the experimental data. Between the wavelengths of 0.3 μm and 1.3 μm the simulation overestimates the total transmission of FTO by 5.8 %.

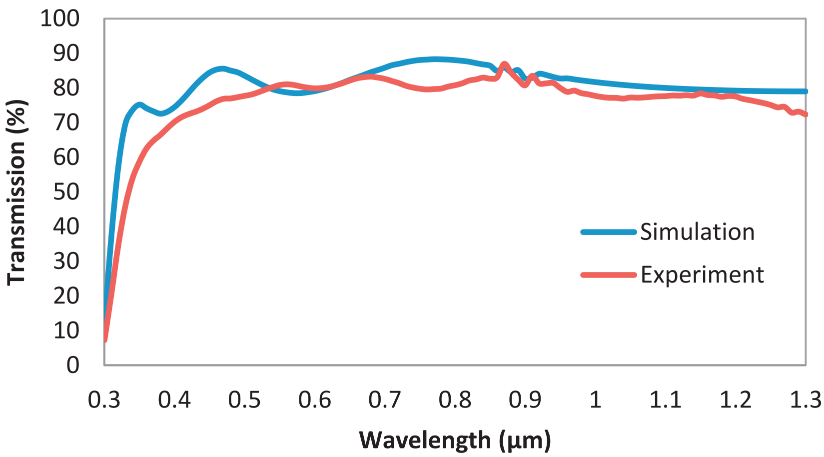


Fig. 3. Transmission curve for 500 nm FTO layer obtained from simulation & experiment.

In order to further validate the simulation, the film's thickness was estimated from the simulated transmission curve using Equations (2) and (3). In the following equations T represents the thickness.

$$T = \frac{\lambda_1 \times \lambda_2}{2\{n(\lambda_1) \times \lambda_2 - n(\lambda_2) \times \lambda_2\}} \tag{2}$$

where λ_1 and λ_2 are the wavelengths of two consecutive peaks and $n(\lambda_1)$ and $n(\lambda_2)$ are the refractive indices of the FTO film at λ_1 and λ_2. Equation (3), from [10], uses a similar approach:

$$T = \frac{\lambda_1 \times \lambda_2}{2(\lambda_2 - \lambda_1) \times n_{av}} \tag{3}$$

where λ_1 and λ_2 are the wavelengths of two consecutive peaks and n_{av} is the average value of the refractive index of FTO in the range λ_1-λ_2.

Table 1 shows the results for Equations (2) and (3) using the transmission curve shown in Figure 3. The results show that the simulation is accurately modeling the thin film characteristics.

Table 1. Thickness predictability of model and empirical equations

Input Thickness in Simulation	Thickness using equation 2	Thickness using equation 3	Percentage of error of predictability of equation 2	Percentage of error of predictability of equation 3
300 nm	341 nm	314 nm	13.6 %	4.67 %
500 nm	460 nm	505 nm	8 %	1 %
900 nm	846 nm	896 nm	6 %	0.44 %

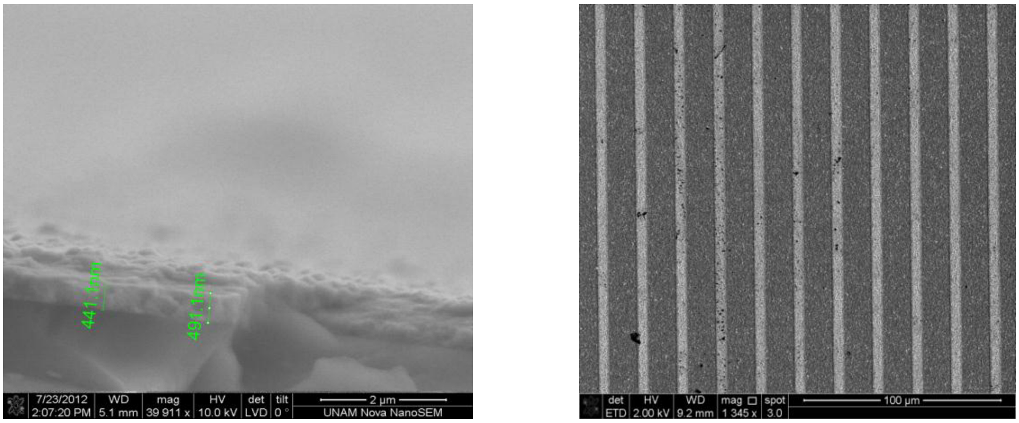
5. Experimental details

Etching experiments were carried out using samples which were provided by an outside group. Each sample consisted of an FTO layer that was 0.5 µm thick on a 3 mm thick layer of glass. The samples were 3 cm by 3 cm. The un-etched samples had an average transmission of 76% in the range 0.3 µm - 1.3 µm as shown in Figure 6. All experiments carried out to date have been based on rectangular etched patterns. The line width, pitch, and etch depth of the pattern were varied.

Prior to the masking step, the sample was cleaned with acetone, isopropanol and deionized water. Samples were then spin coated with the positive resist AZ 1505 and exposed. Etching was carried out using an inductively coupled plasma (ICP) etching machine (LPZ SR (F)). Experiment conditions were chosen as follows: DC bias voltage 270 V, gas composition 20% Cl_2 and 80% Ar with a total flow rate of 25 sccm [8]. After the etching was performed, the samples were cleaned for 20 minutes using O_2 plasma to ash any remaining photoresist.

The physical features of the samples were assessed using a profilometer (P6 Surface profilometer) to measure the etch depth and a scanning electron microscope (FEI Quanta FEG 250) to assess the etch uniformity. The optical characteristics were assessed with a UV-Vis spectrophotometer (Perkin Elmer 1050).

Figure 4(a) shows an SEM image of the cross section of an un-etched FTO sample while Figure 4(b) shows a SEM image of a patterned FTO layer where the bright areas are the un-etched regions. From Figure 4(a) it can be seen that the FTO layer is approximately 0.5 µm thick. The etch rate of the FTO samples was determined to be in the range: 1.5-2.5 nm/sec.



(a) (b)

Fig. 4. SEM image of FTO sample a) Cross section (Operating Voltage 10 kV) b) rectangular patterns (Operating Voltage 2 kV).

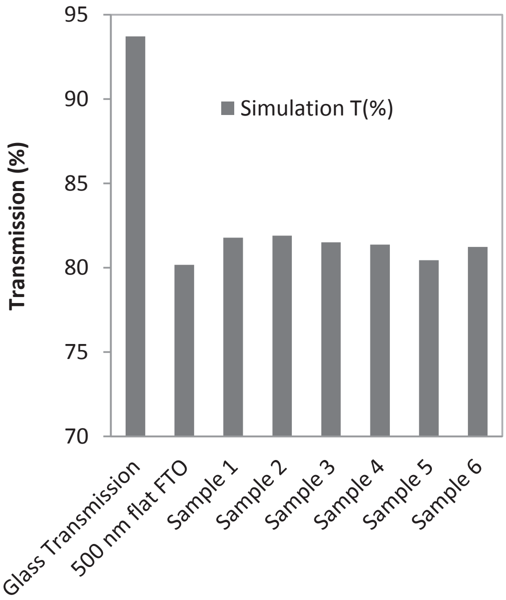
6. Results

Simulations were initially carried out to explore the effect that rectangular patterns would have on the total transmission of the samples. Based on early results, three patterns were chosen for experimental implementation. Once the experiments were carried out, the exact parameters of each sample (as measured after etching) were simulated using the model previously developed. These parameters are shown in Table 2. Simulation results predicted an increase of 1 – 2 percentage points depending on the pattern implemented as shown in Figure 5 (a). These predictions were confirmed by the experimental results as shown in Figure 5 (b).

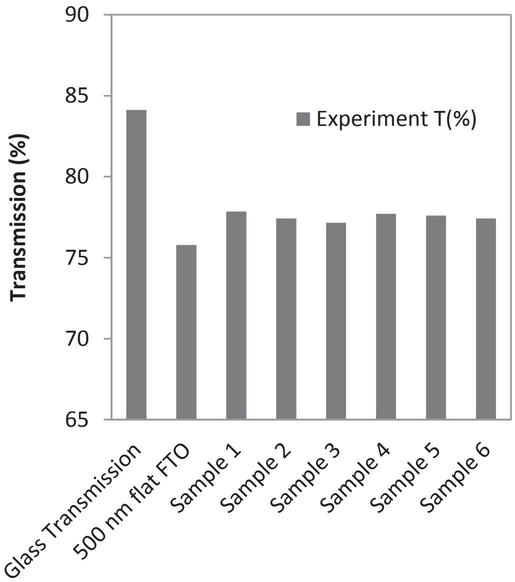
Table 2: Feature size of each sample after ICP-RIE. Experimental conditions were as follows: DC bias voltage: 270 V; gas composition: 20% Cl_2 and 80% Ar; total gas flow rate: 25 sccm)

Name	Width of line (µm)	Pitch (µm)	Etch Depth (nm)
Sample 1	7	30	398
Sample 2	7	30	430
Sample 3	6	20	204
Sample 4	6	20	234
Sample 5	5	24	443
Sample 6	5	24	286

The variation in etch depth is caused by three factors: variation in etch time, a non-optimized recipe, and non-uniformity in the thickness of the supplied samples as shown in fig 4.



(a)



(b)

Fig. 5. Average transmission (%) comparison in between 0.3-1.3 µm range for all the rectangular patterned samples (a) Simulation and (b) Experimental

Further simulation was carried out in an effort to explore the cause of increased transmission. The equivalent thickness of each sample was calculated using Equation (4):

$$T_{eq} = 500 - \frac{ED(P - W)}{P} \tag{4}$$

When ED is the etch depth, P is the pitch, and W is the width, T_{eq} is the uniform thickness a sample would need to have in order to obtain the same volume of FTO as an etched sample with the parameters ED, P, and W. Figure 6 shows the simulation of sample 3 and a structure of equivalent thickness. It clearly shows that the increase in transmission is entirely due to reduced absorption resulting from the decrease in FTO material rather than an increase in light trapping.

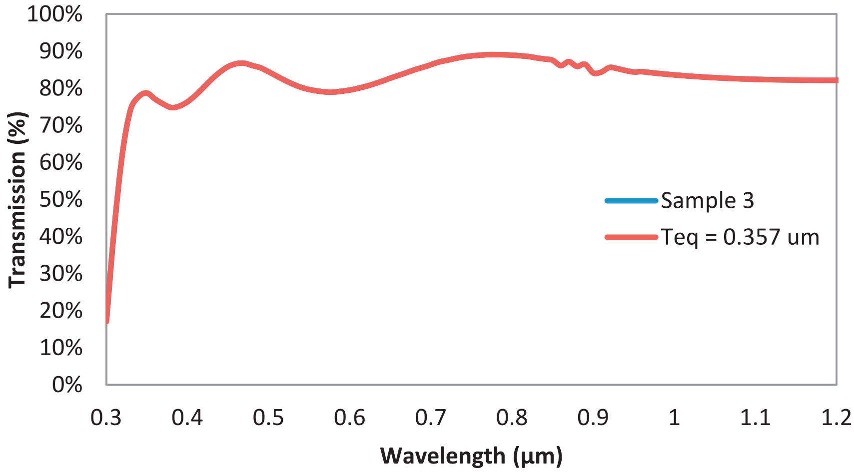


Fig. 6. A comparison between Sample 3 and a sample of equivalent thickness
(The two curves are exactly superposed).

Further simulations were carried out using the triangular patterns. The results are shown in Figure 7. These show that increases in total transmission of over 5 percentage points are possible. Figure 8 shows the optical characteristics for an opening angle of 136°. This clearly shows that the increases in transmission are mostly due to decreases in reflection.

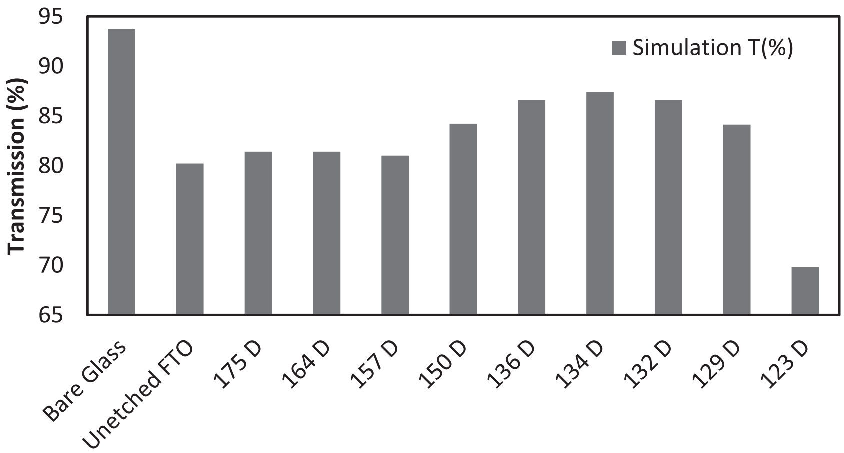


Fig. 7. Average transmission (%) comparison for pyramidal patterned samples (Simulation).

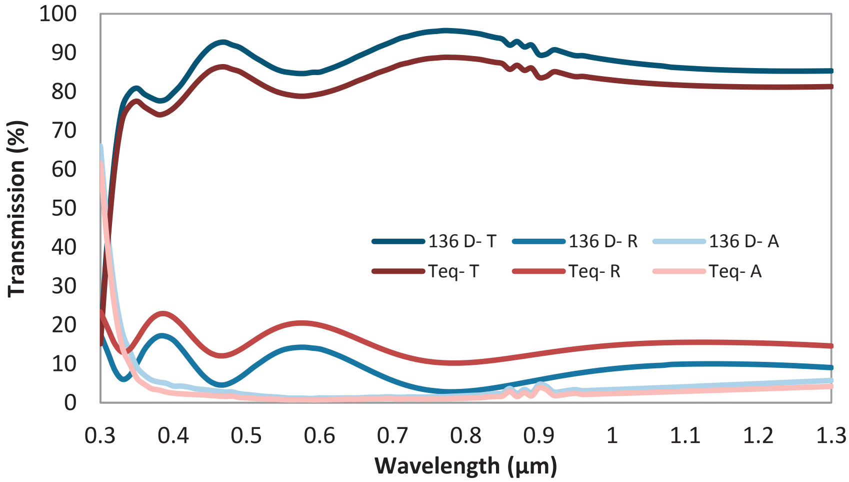


Fig. 8. Average transmission (%) comparison for pyramidal patterned sample with an opening angle of 136° and a sample of equivalent thickness (Simulation).

7. Conclusion

Modern solar cell design critically depends on the development of new architectures for effective light trapping and improving transparent conductive oxide technology. In this paper it was shown that Synopsys TCAD can be used to simulate the optical properties of FTO thin films on glass. The simulation results show that simple rectangular patterns can slightly increase the transmission of samples by up to 2 percentage points but that these gains are entirely due to the reduction in absorption due to the material removed in the etching process. These results were confirmed experimentally. Pyramidal structures were found to have produced transmission increases of over 5 percentage points. Moreover, it was shown that these gains are due to decreases in reflection. Implementing these pyramidal patterns is the subject of on-going research in our group.

References

[1] A. Kumar and C. Zhou, "The Race To Replace Tin-Doped Indium Oxide: Which Material Will Win?," ACS Nano 4, 11-14 (2010).

[2] Brian G. Lewis and David C. Paine, "Applications and Processing of Transparent Conducting Oxides," MRS Bulletin **25**, 22-27 (2000).

[3] Z.Y. Zhong and Y.D. Jiang, "Surface treatments of indium-tin oxide substrates for polymer electroluminescent devices," Phys. Stat. Sol. 203, 3882–3892 (2006).

[4] F. Yang and S.R. Forrest, "Organic Solar Cells Using Transparent SnO2–F Anodes," Advanced Materials 18, 2018-2022 (2006).

[5] T.J. Coutts, J.D. Perkins, D.S. Ginley, and T.O. Mason, "Transparent conducting oxides: status and opportunities in basic research," Electrochemical Soc. Proceeding 99, 274-286 (1999).

[6] J. Muller, G. Schope, O. Kluth, B. Rech, and M. Ruske, "Up scaling of texture-etched zinc oxide substrates for silicon thin film solar cells," Thin Solid Films 392, 327-333 (2001).

[7] TCAD, "Process and device Simulation Tools," http://www.synopsys.com/Tools/TCAD/Pages/default.aspx

[8] Seon Mi Kong, Yubin Xiao, Kyung Ha Kim, Wan In Lee and Chee Won Chung, “Performance improvement of dye-sensitized solar cells by surface patterning of fluorine-doped tin oxide transparent electrodes,” Thin Solid Films 519, 3173–3176 (2011).

[9] J C Manifacier, J Gasiot and J. P. Fillard, “A simple method for the determination of the optical constants n, k and the thickness of a weakly absorbing thin film,” Journal of Phys. 9, 1002–1004 (1976).

[10] F. Demichelis, G. Kaniadakis, A. Tagliaferro and E. Tresso, “New approach to optical analysis of absorbing thin solid films,” Applied Optics 26, 1737–1740 (1987)